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**6. AUTHOR(S)**
DR. LAWSON

**7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES)**
THE UNIVERSITY OF ALABAMA
IN BIRMINGHAM
701 20TH STREET SOUTH
BIRMINGHAM AL 35294-0111

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Final Report

## Optical Low Coherence Reflectometry Based On Degenerate Four Wave Mixing In Thin Films For Non-Invasive Studies of Air-Craft Composite Materials

Chris M. Lawson, University of Alabama at Birmingham


### Project Abstract

Optical coherence-based imaging techniques have recently been studied[1] for non-destructive characterization of diffusive materials such as composites, polymers, and coatings used in aircraft components and systems. However, optical photons are strongly scattered in these diffusive materials, and this scattering process can severely degrade the optical image resolution by randomizing the information in the transmitted or reflected optical signals, so to achieve good optical image resolution it is necessary to differentiate between image-bearing and strongly scattered photons. A number of single-point Optical Low Coherence Reflectometry (OLCR) techniques have attracted attention for these applications, but these single-point OLCR techniques require transverse scanning of the sensor head, which can lead to long image acquisition times. Conversely, real-time holographic methods in photorefractive materials can provide cross-sectional images (at a given depth) in scattering media with no sensor head scanning. However, the depth resolution and field-of-view (FOV) in these previous photorefractive-crystal based imagers have been limited by beam-walk-off effects, where the depth corresponding to equal optical path lengths in the two writing beams varies across the beam intersection plane.

We propose a novel approach to significantly improve the spatial resolution and FOV of these real-time holography-based imagers. We propose a degenerate four wave mixing (DFWM)-based imager, with the photorefractive crystal replaced by a thin, highly nonlinear optical (NLO) liquid crystal film. Our imager provides instantaneous two-dimensional (2-D) cross-sectional images (depth scanning gives the third dimension) without the need for transverse scanning of the sensor head. In addition, our DFWM process in thin dye-doped liquid crystal films enables very small beam crossing angles that minimize beam walk-off effects, and will lead to significantly improved depth resolution and FOV. When used in DFWM configurations, our liquid crystal doped dye films have exhibited: (i) strong orientational photorefractive effects with small beam crossing angles (which minimizes beam-walk off effects); (ii) strong phase conjugate reflectivities (which gives good imaging sensitivities) for low intensities; (iii) low cost; and (iv) significantly improved the NLO performance with the application of an external electric field. We will use ultra-short Ti:sapphire laser pulses, which have a high degree of spatial coherence but have a very short coherence length, as required, and later we will frequency double these pulses to enhance the resolution (because of the shorter wavelength).

The development of these high resolution, DFWM-based imaging techniques for providing single-pulse cross-sectional images versus depth will have intrinsic scientific value, as well as, a number of important applications for the Air Force. These Air Force applications include non-destructive evaluation of the porous structure of transparent, but highly diffusive, epoxy composites, as well as, characterization of polymers and thin film coatings used for Air Force components and systems.

## PROJECT DESCRIPTION

### A. Program Background And Significance

There are a number of important commercial and Department of Defense (DoD) applications that require non-invasive imaging of the internal structure of diffusively scattering materials. One application of relevance to the Air Force involves non-invasive imaging of the internal porous structure of certain types of transparent (but diffusive) composite materials.[1] The multiple scattering process in these diffusively scattering materials can severely degrade image resolution by randomizing the optical image information in the transmitted or reflected optical signals. To achieve good optical image resolution in these inhomogeneous materials, it is necessary to differentiate between minimally scattered image-bearing photons and those which have been scattered many times, and numerous time domain and frequency domain filtering techniques have been used for this purpose.[2,3,4,5,6,7,8,9,10] Optical low coherence reflectometry (OLCR) has been one of the most popular techniques for these applications. At UAB, we have investigated the use of fiber-based OLCR techniques for a number of years for non-invasive studies of polycrystalline solar silicon,[11,12,13,14,15] with approximately a 1 μm resolution. More recently, we have used these techniques for providing non-invasive images of protein crystals in solution.[16]

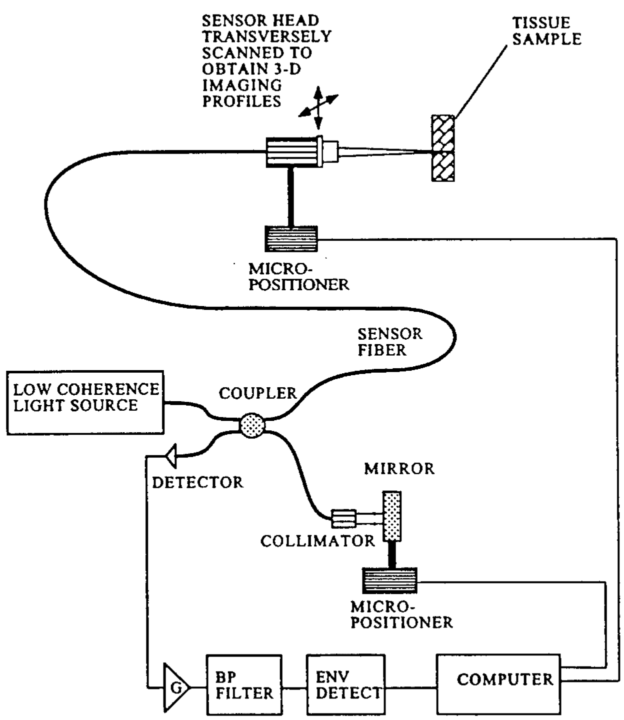

*Figure 1 Single Point OLCR Sensor System*

The operation of one of our ***single-point*** OLCR systems is shown in Figure 1, with one of our example images (of a lysozyme crystal) from this system shown in Figure 2.[16] Broad spectrum light is directed into a single mode fiber and coupled into the sensor leg and reference leg of a fiber optic Michelson interferometer. Light that exits the sensor fiber is collimated and backscattered from a sample, re-enters the fiber, and is re-combined with light from the scanning reference leg. Strongly modulated interference fringes only occur when the optical path length difference between the sensing leg and the

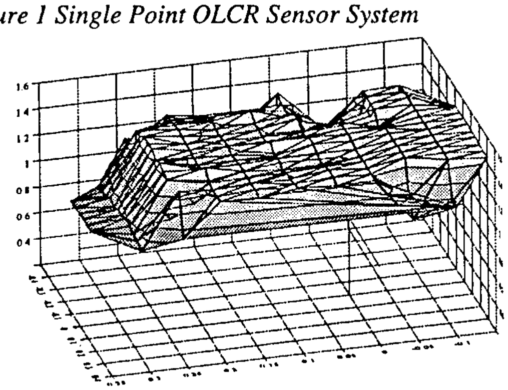
*Figure 2 Example of transversely scanned single-point OLCR image*

reference leg is less than $L_c$, the short coherence length of the broad spectrum light source. Hence, fringes are observed when the reference leg optical path length is equal to the sensor leg optical path length established by backscatter from some feature of interest. By monitoring the absolute position of the reference leg mirror that corresponds to these "peaks" in oscillating interference fringes, one obtains a direct, high resolution, absolute measurement of the optical backscatter vs. depth. Full three-dimensional (3-D) images like that shown in Figure 2 are then obtained by transversely scanning the sensor head in the two directions orthogonal the optical axis. *A major disadvantage of these single-point OLCR systems is that the sensor head must be transversely scanned*, which can lead to long image acquisition times.

An alternative approach for imaging through inhomogeneous media which eliminates this transverse scanning requirement involves the use of holographic techniques,[17] as well as, various types of pulse gating techniques based on the use of NLO materials, including stimulated Raman amplification,[18] parametric image amplification,[19] and second harmonic up-conversion.[20,21] Recently, two-wave mixing in photorefractive crystals (rhodium-doped barium titanate) has been used for holographic based imaging through a turbid media.[22,23,24] However, photorefractive crystals can be expensive and can be susceptible to large beam-fanning effects[25,26] that serve as a noise source for the imaging process. Also, *the image resolution and field-of-view (FOV) for coherence-based depth resolved imagers based on photorefractive crystals are limited by beam-walk-off effects*[23,27] because of the large beam crossing angles that are required in these photorefractive-crystal based systems. This means that a coherence-based depth-resolved image of the object is narrowed as shown in Figure 3,[28] which shows a resolution chart image recorded in a 1 mm thick Ba:$TiO_3$ crystal with mode locked transform limited laser pulses, with a beam crossing angle of 9 degrees. For comparison purposes, a non-depth-resolved reference hologram is shown in Figure 4 (with a high coherence length He:Ne laser source). It can be seen that the FOV of the coherence-based depth resolved image we obtained with a photorefractive crystal in Figure 3 is significantly reduced compared to the full image.

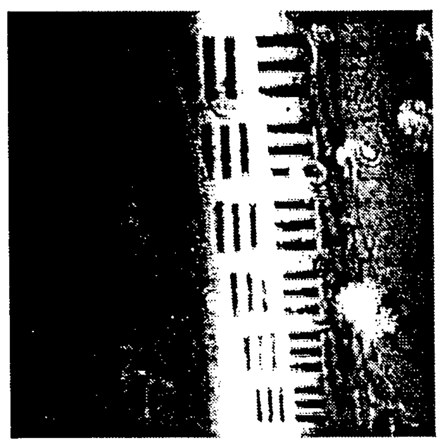

*Figure 3 Illustration of Beam-Walk-Off effects in* ***coherence-based depth-resolved*** *2-D imaging. Resolution chart image obtained via DWFM on Ba:$TiO_3$ crystal with Ti:Sapphire laser (130 fs) pulses.*

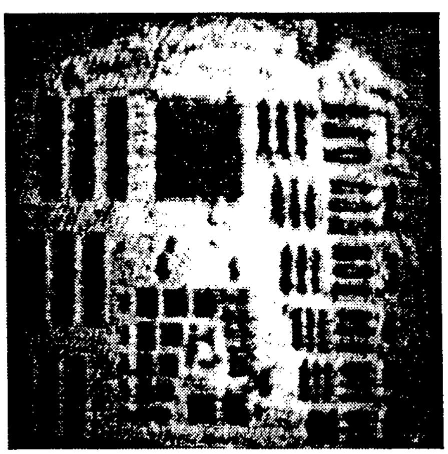

*Figure 4 Reference hologram of chart* ***(no coherence-based depth resolved imaging technique used).***

The origin of beam-walk-off is illustrated in Figure 5. When a short coherence length light source is used for nonlinear wave mixing in a photorefractive (as is required for *depth resolved* coherence imaging techniques),[22-24] the working volume of the "NLO filter" is significantly reduced and can become much smaller than the beam overlap region. This phenomena occurs because two interacting beams will be coherent only in a narrow area of their intersection which is determined by their angular separation. As a result, the FOV is decreased due to the angular separation between the writing and the reference beam.

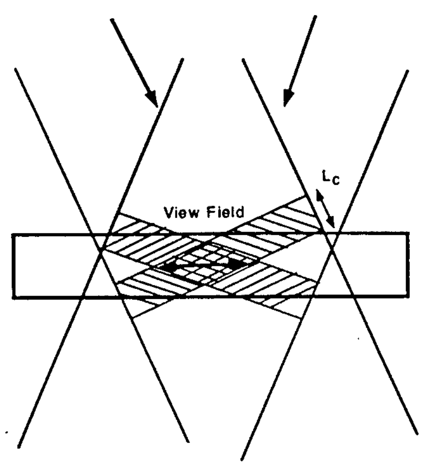


*Figure 5 Illustration of Beam-Walk-Off Effects. Overlapping area within coherence length $L_c$ of the probe and forward pump beams is decreased with large beam separation angle, α.*

It should be noted that because of the short coherence length of the light source, only a thin layer of the photorefractive material (comparable to $L_c$) can be efficiently 'used' as an active volume for the NLO wave mixing-based coherence filtering. The "unused" volume of the photorefractive exposed to the beams will only increase undesirable beam fanning effects.

The way to provide a large imaging field is to decrease the beam crossing angle, α. As one can see from Figure 6, it is not practical to significantly decrease α in photorefractive crystals (such as $Ba{:}TiO_3$), because the maximum of the conjugate reflectivity $R_c$ appears at $\alpha \sim 45^o$, and $R_c$ decreases strongly with decreasing α. We have recently established this[29] in measurements performed on $45^o$-cut $Ba{:}Ti0_3$ crystal (which has better performances at small angles than a $0^o$ -cut crystal). Our measurements were performed with a 5 mW He:Ne laser light source. As can be seen from our data, at $\alpha=5^o$ the phase conjugate reflectivity $R_c$ has decreased by an order of magnitude. Hence, this data illustrates that if one attempts to minimize beam-walk-off effects by minimizing the α in photorefractive crystals, there are significant decreases in the phase conjugate reflectivity, which will decrease the sensitivity of the imager. In order to solve this problem, one must find thin NLO materials that exhibit high phase conjugate reflectivities at small beam crossing angles. Multiple quantum well structures[30] and photorefractive films[31] have recently been studied for these applications.

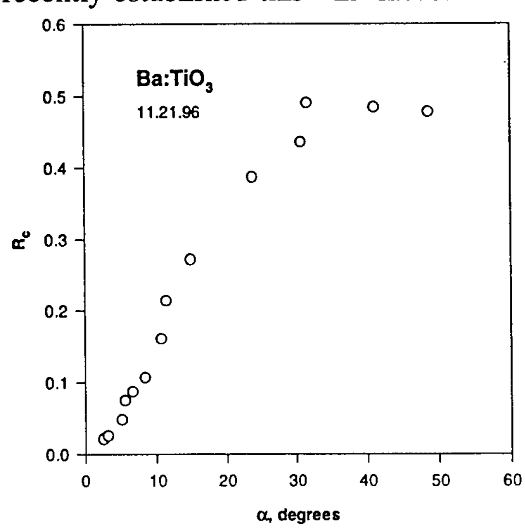


*Figure 6 Phase conjugate signal $R_c$ of $45^o$-cut $Ba{:}TiO_3$ crystal versus crossing angle between the probe and forward pump beam, This data shows that $R_c$ decreases with decreasing angles in this photorefractive crystal.*

We have recently studied nematic liquid crystal films for this application. When these films are used in a degenerate four wave mixing (DFWM) geometry, the maximum of diffraction efficiency (and, hence, the phase conjugate reflectivity) is observed at significantly smaller crossing angles $\alpha$, than for the $Ba:TiO_3$ photorefractive crystals.

Figure 7 illustrates the angular behavior of the phase conjugate reflectivity of a 100 µm-thick LC film with a Q-switched 150 mW Alexandrite laser light source at 20 Hz repetition rate. It can be seen that there is a sharp increase in the phase conjugate reflectivity at angles smaller than $8^{o}$.

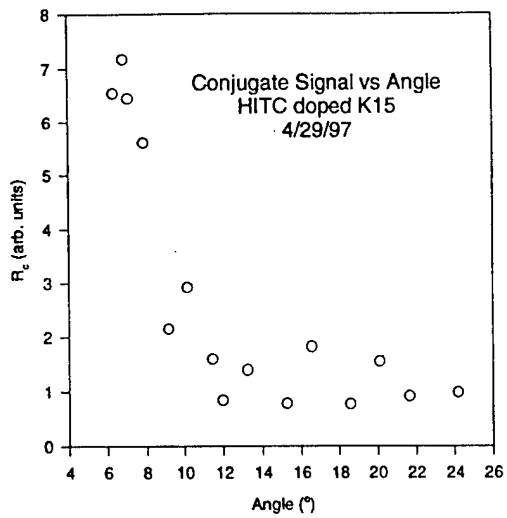


*Figure 7 Phase conjugate signal $R_c$ vs. probe-pump crossing angle, $\alpha$, for DFWM in a 100µm dye-doped LC film. $R_c$* ***increases*** *as the $\alpha$ decreases, so that small crossing angles can be used (minimizing beam-walk off effects).*

## B. Program Objective

We propose a novel approach to significantly improve the spatial resolution and FOV of real-time holography based imagers. Instead of two-wave mixing in a thick photorefractive crystal, we propose a degenerate four wave mixing (DFWM)-based imager, with the photorefractive crystal replaced by a thin, highly nonlinear optical (NLO) nematic liquid crystal film. Our imager provides *instantaneous two-dimensional (2-D) cross-sectional images (depth scanning gives the third dimension) without the need for transverse scanning of the sensor head. Furthermore, our DFWM process in thin dye-doped liquid crystal films allows for the use of very small beam crossing angles that minimize beam walk-off effects, and will lead to significantly improved depth resolution and FOV.* Our NLO films will be fabricated by confining dye doped liquid crystal solutions between fused quartz plates. Additional performance enhancements can be obtained by applying an external electric field across the liquid crystals.

Initially, we will be using ps and fs Ti:sapphire laser pulses, which have a high degree of spatial coherence but have a very short coherence length, as required. Later, we will extend the types of low coherence light sources used for this application by using frequency doubled Ti:sapphire laser pulses, which have the potential of enhancing the resolution because of their shorter wavelength.

The development of this high resolution, DFWM-based imaging technique for providing single-pulse cross-sectional images versus depth will have an intrinsic scientific value, as well as, a number of important applications for the Air Force. These Air Force applications include non-destructive evaluation of the porous structure of transparent, but highly diffusive, epoxy composites,[1] as well as, characterization of polymers and thin film coatings used for Air Force components and systems.

## C. Experimental Methods and Procedures

In this section, we will describe some of our proposed experimental methods and procedures, along with some preliminary results. Section C.1 describes our experimental technique for obtaining single-pulse 2-D cross sectional images (which can be depth-scanned to yield the third dimension) with some example images. Section C.2 describes preliminary results which investigate the spatial resolution of the technique, and Section C.3 describes proof-of-principle experiments where we have successfully demonstrated instantaneous 2-D cross-sectional imaging in scattering media, using DFWM in thin NLO films made of dye-doped liquid crystals. Section C.4 describes experiments where we have explored the limits of signal detection in very highly turbid media. Finally, Section C.5 describes our proposed techniques for improving the sensitivity and resolution of the imager.

### 1. Proposed UAB Instantaneous 2-D Imaging Research Efforts and Preliminary Results

Our proposed technique, described in detail elsewhere,[28,32,33] is based on degenerate four wave mixing (DFWM) in a thin nonlinear optical liquid, and it provides single pulse 2-D image cross-sections at a given depth without sensor head scanning. Our experimental configuration, shown in Figure 8, utilizes a broadband laser source in a DFWM configuration with a CCD camera-based imaging system. The sample is placed in a probe beam optical delay line. Coherence gating of 2-D images is accomplished via DFWM on a grating created on a thin NLO film. Backscattered light from different depths of the sample form an "object" beam, which serve as a probe beam in DFWM scheme. The phase conjugate signal from the DFWM process is only formed whenever the difference in the light paths from the "object" beam (for a given depth) and the forward pump beam is less than $L_c$, which is very small for the broadband

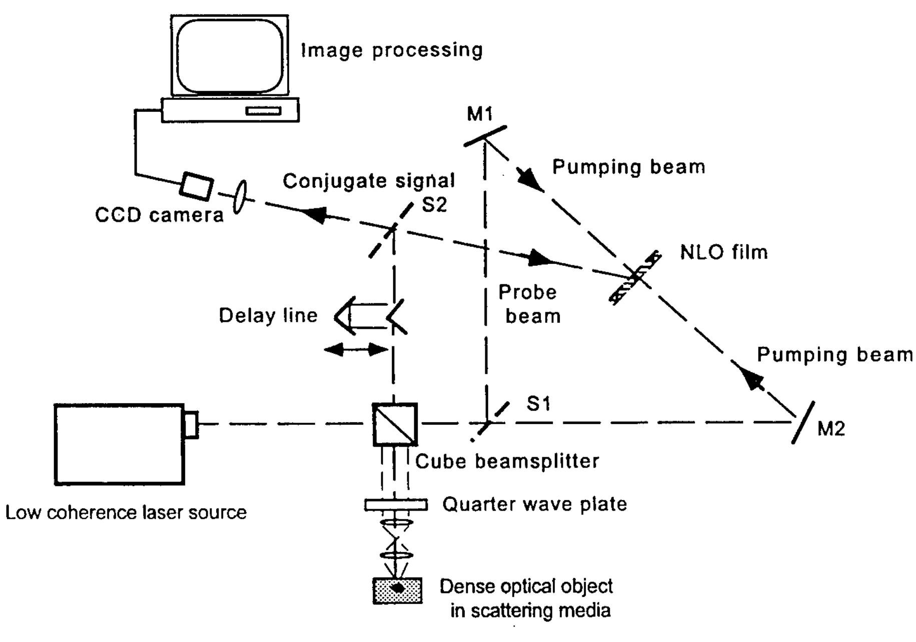


*Figure 8 Instantaneous 2-D Imaging Optical Set-Up*

source. The depth profile of the sample can be scanned by varying the optical delay path, and the two-dimensional profile is instantaneously imaged in real time with the CCD camera.

Preliminary proof-of-principle experiments were performed with an alexandrite laser (Light Age, Inc.) light source at 750 nm. This laser provides a relatively short coherence length of about 5 mm when it operates in Q-switch regime (60 ns pulse width). The angle between the probe beam and forward pump beam (See Figure 8) for the 2-D imaging experiments was set to approximately 6°. The translation stage, located behind the quarter wave plate, served as an optical delay line. Mounted on the translation stage was a letter "A" (approximate size of 3 mm) drawn on a microscope slide. A COHU 4800 CCD camera was used to register the conjugate signal of the image of reflected (back scattered) light from the object.

As a real time holographic recording medium we used a thin NLO layer of nematic liquid crystal E7 (provided by EM Industries, Inc.) which exhibited strong optically induced refractive index changes. The sensitivity of the thermal component to these optical index changes was enhanced by the addition of an infrared (IR) dye[34] (BDN, provided by Exciton Inc.). The liquid crystal layer, which had a thickness of 100 μm, was arranged between two microscope slides. To obtain planar alignment of the nematic phase liquid crystal, the interior surfaces of the glass slides were coated with polyvinyl alcohol and unidirectionally buffed.

*Figure 9 Instantaneous 2-D Image On CCD Array When Depth Is Tuned To The Object (Letter "A") Depth. Image Is Clearly Visible.*

Examples of 2-D images obtained by the CCD camera using this technique are shown in Figure 9 and Figure 10, where the "dense optical object" being imaged was a letter "A" in front of a retroreflector. Figure 9 shows a clear CCD camera image of the "A" when the depth established by the delay line is tuned to the depth of the "A", and Figure 10 shows that no image is observed when the depth is tuned off of the depth of the object.[33] The technique that we have described here and in Reference 33 has the advantage that a variety of low coherence light source types can be used. In addition, each 2-D image (for a given depth) can be acquired in a single laser pulse, which is particularly appropriate for applications such as composite evaluation where cost constraints may preclude long horizontal scanning times.

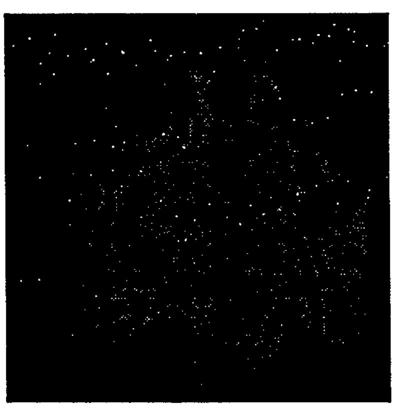

*Figure 10 Instantaneous 2-D Image When Depth Is Tuned Off Of Object (Letter "A") Depth. No Image Is Visible On CCD Array.*

2. Spatial Resolution Experiments and Preliminary Results

*Figure 11 Two-dimensional image of Air Force resolution chart; finest bars correspond to 14.3 LP/mm. The angle between the probe and pump beam was approximately 6°.*

The transverse resolution of our imaging technique can be improved by improving the mode quality of the laser beam and by improving the resolution of the image capture technique. Preliminary experiments to improve spatial resolution involved the introduction of an intra-cavity aperture to improve the mode quality of the laser beam, as well as, using a frame grabber card with 512 X 512 pixel resolution. A 125 cm focal length lens was inserted into the probe beam arm of the DFWM setup to relay the image into the dye-doped liquid crystal with 1.3 times demagnification.

To evaluate the achievable resolution in the transverse plane with dyed doped liquid crystal films as the holographic recording media, we performed imaging experiments with an Air Force resolution chart as a test object. Our optical pump source was a Q-switched Alexandrite laser operating at 20-Hz. The result shown in Figure 11 is an image of the resolution chart taken with a *single shot* of the laser pulse and demonstrates our achievable resolution of 70 $\mu$m at $\lambda$=740 nm. These results show that we have already obtained excellent spatial resolution, and as described in Section C.5 we should be able to improve this resolution in the future by improvements in the transverse mode quality and stability of the laser source.

3. Imaging Experiments In Turbid Media

Investigations of the 2-D imaging resolution that can be obtained through a turbid media can be performed by inserting a diffusive scatterer into the experimental configuration of Figure 8. Preliminary experiments were performed with a resolution chart as a test object and a suspension of polystyrene microspheres in water as a scattering media. Figure 12 shows an image acquired with a single shot of the laser beam without a scattering media present. The bars correspond to 1.41 LP/mm (about 700 $\mu$m) on the resolution chart. The NLO media used for this experiment was K15 liquid crystal (EM Industries) doped with an infrared dye (HITC from Exciton), with an absorption of 1.03 at the working wavelength (740 nm). The ratio of the intensities of the probe beam to the forward and backward pump beams was 0.67:1:1, and the laser beam spot size on the liquid crystal surface was about 3.5 mm . Figure 13 corresponds to a scattering media with 0.027 % concentration (0.9 mean free paths) of polystyrene microspheres in water, and Figure 14 corresponds to a scattering media with 0.04% concentration (1.3 mean free paths).

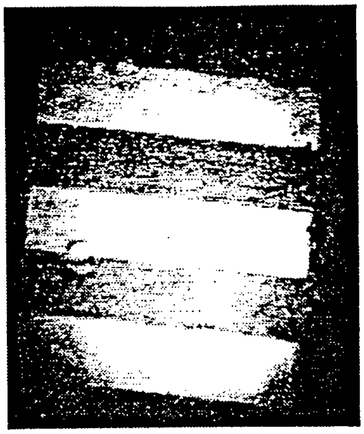

*Figure 12 Image of a section of the Air Force resolution chart which corresponds to 1.41 LP/mm, No scattering media*

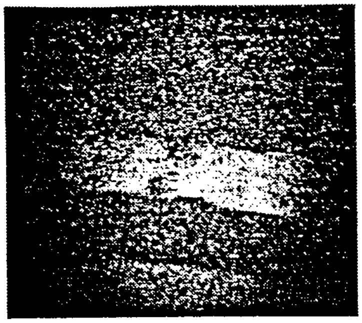

*Figure 13 Image of Air Force resolution chart with scattering media of .02% concentration.*

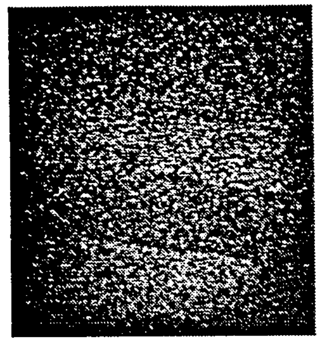

*Figure 14 Image of Air Force resolution chart with scattering media of .04% concentration*

As can be see from the images in Figure 13 and Figure 14, we have clearly demonstrated instantaneous (non-scanning) cross-sectional imaging in a turbid media using our technique. *These results are significant in that they represent the first demonstration of this technique that we are aware of to provide instantaneous (non-scanning) cross-sectional images in a turbid media.* In the next section, we will show some preliminary results where we have demonstrated very sensitive 1-D imaging through a very highly diffusive media, and we will subsequently discuss proposed techniques for improving the sensitivity of the 3-D imaging method.

4. 1-D High-Sensitivity Experiments Through Very Turbid Media

We have also performed proof-of-principle experiments to demonstrate the applicability of our coherence filtering technique in a thin NLO material for high-sensitivity 1-D imaging through a very highly scattering media. The DFWM configuration, described in detail elsewhere[32,33], is similar to that illustrated in Figure 8, but with a single detector used instead of a CCD detector array.

A frequency doubled LiF:$F_2^-$ color center laser (CCL)[35] pumped by a Q-switched, 10 Hz repetition rate Nd:YAG laser was used as the light source for our experiments. The CCL output was frequency doubled with a $LiIO_3$ crystal without beam focusing. The doubled laser spectral output was centered at $\lambda$ = 575 nm with a spectral width $\Delta\lambda$ = 0.67 nm, which yields a temporal coherence length $L_c \sim \lambda^2/\Delta\lambda \sim 490$ µm. The typical CCL pulse energy was 0.1-0.5 mJ, and the pulse duration was about 4 ns.

The coherence filtering operation was performed by DFWM in a thin layer of dye solution in a 100-µm thick optical cell. As a nonlinear optical (NLO) media, we used a solution of Disperse Red 1 in $CH_2Cl_2$ at a concentration of 1.4 mg/ml, which exhibits strong thermally induced refractive index changes.[36] For the working wavelength $\lambda$=575 nm the absorption value in the dye solution was ~ 0.4. To increase the intensity of

conjugate signal, we place a 75 cm lens at the entrance of the DFWM scheme to focus the probe and pump beams on the NLO sample. The magnitude of the phase conjugate reflectivity was about 1%.

The probe beam was directed to a 'hidden sample' - a flat object mirror $M_1$ placed behind a scattering cell containing a suspension of polystyrene microspheres in water solution. The object mirror $M_1$ is mounted on a translation stage that functions as an optical delay line. The angle between the probe beam and the forward pump beam was approximately 13°. Detectors (Molectron J3S-10 and J4-09 energy meters) were used to detect the phase conjugate signal and to provide a reference pulse energy monitor, respectively.

The backward pump beam in that experiment was orthogonally polarized in respect to the other two beams with the help of a half-wave plate. As a result, the phase conjugate signal is generated via diffraction of the backward pump beam on the static phase grating formed by the interference of the probe beam and forward pump beam[37] with a grating period $\Lambda=\lambda/[2\sin(\alpha)] \cong 2.2\ \mu m$. The polarization of the conjugate signal in our case is parallel to the polarization of the backward beam.

Figure 15 shows experimental results which demonstrate our approach for low-coherence reflectometry with DFWM coherence filtering in a thin NLO layer. Good fringe visibility for the phase grating is only obtained when the optical path length mismatch (for a given position of the delay line) between the forward pump and probe beams is less than the coherence length of the light source. Figure 15a) illustrates an autocorrelation trace for DFWM measurements with low-

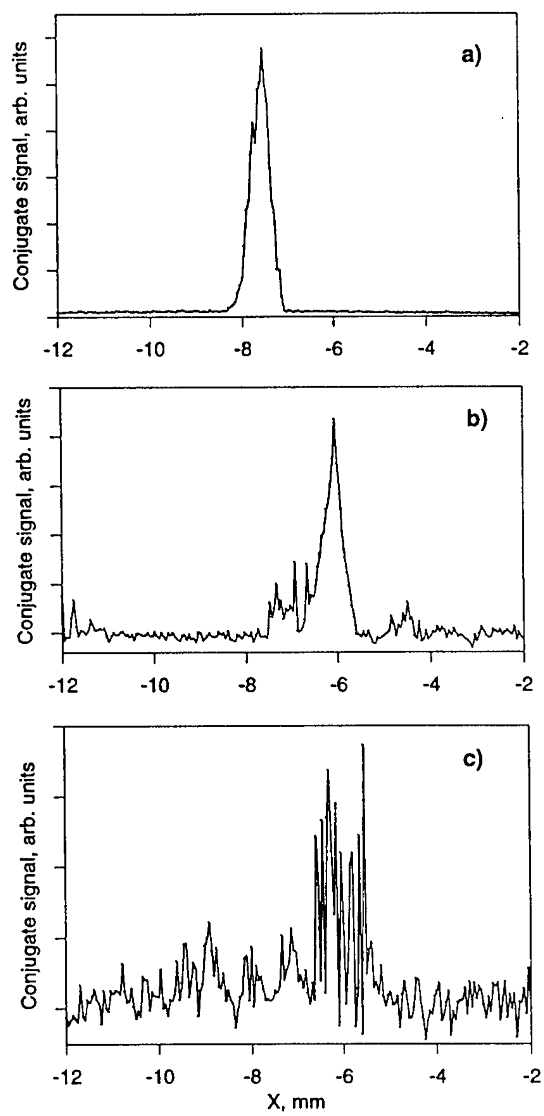


*Figure 15 Output signal with: a) - no scattering cell, peak power of the laser pulses $W_p$= 29 kW; b) - scattering cell with 0.07% concentration 1.0 μm polystyrene spheres in water, $W_p$= 42 kW (6 mean free paths); c) - scattering cell with 0.3% concentration 0.46 μm polystyrene spheres in water $W_p$= 36 kW (14 mean free paths).*

coherence nanosecond laser pulses without the scattering media in the probe beam arm. This plot shows that the full width at half maximum of the phase conjugate signal is about 470 μm. This spatial width is larger than the thickness of the NLO material and is comparable with the estimations for $L_c$ obtained from spectroscopic measurements. The peak in Figure 15a) shows that the position of the object mirror $M_1$ can be measured with an accuracy of about 100 μm. The use of our DFWM filtering technique to detect the reflected signal from the object mirror when the probe beam propagates through turbid media is shown in Figure 15b) and c), where c) corresponds to higher scattering conditions than b). In these experiments, the scattering conditions were provided by a suspension of polystyrene microspheres in water in a 1-mm thick optical cell. For the case shown in Figure 15b), the scattering cell contained 1 mm diameter polystyrene microspheres at a concentration of 0.07% (in accordance with Mie scattering calculations[38] these conditions correspond to about 6 mean free paths in the double pass through the scattering media).

The level of the signal registered by the detector for the case shown in Figure 15b) was more than two orders of magnitude lower than without the scattering cell, and it contained strong background noise. To increase the sensitivity of the method for each position of the delay line, the average value of the conjugate signal $\langle E_C \rangle = \langle E_{D1} - K*E_{D2} \rangle$ was calculated after every laser shot, where $E_{D1}$ and $E_{D2}$ are the energy of the pulses registered by detectors $D_1$ and $D_2$, and K is a coefficient ($K \sim 10^6$) characterized by the ratio of the energy of the conjugate signal to the pump energy.

Despite the very large increase in the optical background level due to scattering from the diffusive media, a signal from the object mirror can clearly be resolved (with about two orders of magnitude attenuation compared to that without the cell). As it can be seen from Figure 15b), the maximum of the phase conjugate signal is shifted by 1500 μm to the right when compared with the maximum of Figure 15a), because of the increase in the optical path length of the probe beam arm introduced by the presence of the scattering cell. The conjugate signal formed by the scattered probe beam in the case shown in b) has a sharply peaked maximum, and the position of the mirror can be determined with about the same 100-μm accuracy as for the measurement without the scattering media. In the case of Figure 15c), the strongly scattering cell contained 0.46 mm diameter polystyrene microspheres at 0.3% concentration, corresponding to 14 mean free paths in a double pass. Because this cell had much higher scattering than that shown in b), leading to a higher probe beam attenuation, the magnitude of the phase conjugate signal was more than an order of magnitude lower than that shown in b). For the strong scattering case shown in Figure 15c), the decreased signal to noise ratio (SNR) reduces the accuracy with which the position of the mirror can be determined to about 1 mm. It is important to note that the operational peak power of the nanosecond pump pulses for the coherence filtering through the scattering media in our experiments was about 40 kW which corresponds to about 1.6 mW of average power for the CCL and is comparable with the average power of superluminiscent light emitting diodes.

*In summary, in this section (C.4) we have described experiments to explore the limits of signal detection using this technique in very highly scattering media. In the previous section (C.3) we were able to clearly demonstrate the feasibility of using DFWM-based coherence imaging in thin NLO films for imaging in scattering media, which represent the first demonstration of this technique to provide instantaneous (non-scanning) cross-sectional images in a turbid media.*

5. Proposed Techniques for Improving Sensitivity and Resolution

Although we have already obtained proof-of-principle preliminary results which demonstrate the very promising potential of our imaging technique, there are a number of modifications that we plan to make in the proposed program to improve signal to noise ratio (SNR) of our images, and thus the sensitivity of our imaging technique. In our future experiments, instead of a Q-switched laser source we are going to use a quasi-CW mode-locked Ti:Sapphire laser as a low-coherence light source. This laser generates a transform limited pulse train with about a 100 femtosecond pulse width. The transverse mode quality and stability of this quasi-cw mode-locked Ti:sapphire laser will be much better than that we obtained with the alexandrite laser, and this will improve the transverse resolution of the images obtained with our technique. In addition, when transform-limited pulses are used, the coherence filtering process via DFWM on the NLO material can be also considered as a pulse gating technique.[39] Because of a very short temporal width of the laser pulses, the arrival time of the ballistic photons (image bearing photons which are backscattered from the target) can be separated in time from the moment of arrival of the more numerous "noise background tail" photons formed by the randomly scattered photons in a turbid media. Hence, in the wave-mixing process in the NLO sample, the image bearing photons will not overlap with the noise photons, improving the sensitivity (i.e., the SNR) of the method.[39] Later, we will extend the types of low coherence light sources used for this application by using frequency doubled Ti:sapphire laser pulses which have the potential of yielding resolution enhancements because of their shorter wavelength.

However, to realize these expected improvements in spatial resolution, as a practical matter it is also necessary to take into account beam geometry effects, which can lead to very significant FOV limitations in photorefractive-crystal based imaging configurations, due to beam walk-off effects. As we described in Section A, we have shown that if nematic liquid crystal films are used in a degenerate four wave mixing (DFWM) geometry, $R_c$ actually increases as the angle gets smaller (see Figure 6); hence, we propose to use very small beam crossing angle configurations to minimize these beam walk-off effects.

The use of the quasi-CW mode-locked Ti:sapphire laser will also improve the sensitivity because it will enable us to use NLO materials with stronger, slower nonlinearities, and accumulate the NLO response in a manner similar to that used with photorefractive crystals. In our previous experiments using liquid crystal films we utilized a thermal mechanism of nonlinearity[40] which requires higher intensities than for phase

conjugation with photorefractive crystals, such as Ba:$TiO_3$. We propose to obtain significant NLO sensitivity improvements (i.e., improvements in the phase conjugate reflectivity of the DFWM process) via laser-induced molecular reorientational effects in dye doped nematic liquid crystal films by applying external DC electric fields.[40,41,42,43,44,45]

With these DC fields applied, the optical intensity required for nonlinear wave mixing is about 0.2 W/cm$^2$,[43] which is comparable with the intensities required for the wave mixing in photorefractive crystals (without the DC field applied to the liquid crystals, the optical intensity needed to generate similar diffraction efficiency is about 6W/cm$^2$). The maximum diffraction efficiency (at small angles ~ 2-3°) for liquid crystal films can be as high as 10-20%,[40,43] which is much larger than the diffraction efficiencies of photorefractive crystals at small angles (See Figure 6).

We have performed some very preliminary DFWM experiments to verify improvements in the NLO response of our dye doped nematic liquid crystal films that can be obtained via laser-induced molecular reorientational effects when an external DC electric field is applied. Figure 16 shows the dependence of the phase conjugate reflectivity at low beam powers versus applied voltage for a K15 liquid crystal (EM Industries) doped with an infrared dye (HITC from Exciton, Inc.). The thickness of the liquid crystal layer is about 95 μm, and the absorption at the working wavelength (780 nm) was about 0.076, and the probe beam intensity was about 700 mW/cm$^2$. It is important to note that this data was obtained with a small (about 3 degrees) crossing angle between the probe and forward pump beams, as is required for minimizing beam walk-off. It can be seen from Figure 16 that the phase conjugate reflectivity for this initial experiment is increased from a

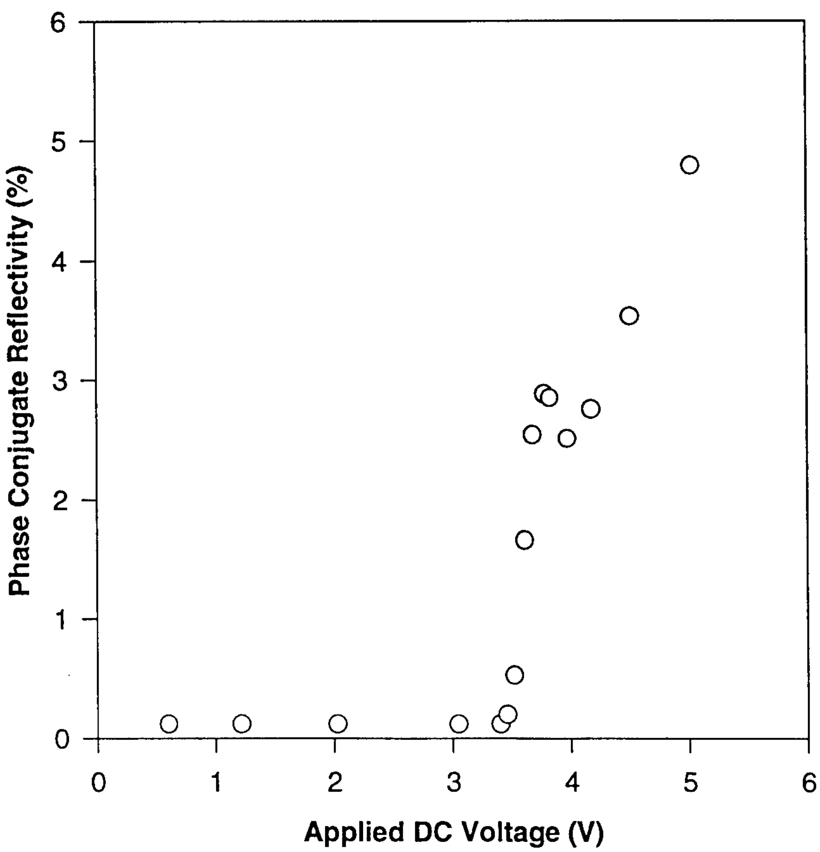


*Figure 16 Phase conjugate reflectivity vs. applied DC voltage for HITC doped K15*

negligible value to almost **5%** as the voltage is increased from 0 to 5 volts because of the Friedricks-transition effect.[40-45] We anticipate further improvements in the nonlinear performance of the medium as we optimize experimental parameters such as the type of liquid crystal-based material and the beam crossing geometry.

In summary, in our initial proof-of-principle experiments we have successfully demonstrated instantaneous 2-D cross-sectional imaging in scattering media, using DFWM in a thin NLO films made of dye-doped liquid crystals. These dye-doped liquid crystals have exhibited very strong thermal and orientational optical nonlinearities.[40-45] We propose to significantly improve this technique using a quasi-cw mode-locked Ti:Sapphire laser with Fourier-transform limited femtosecond pulses as a low coherence source. The liquid crystal films discussed above are very attractive for imaging applications via NLO filtering because: (i) they show strong orientational photorefractive effects under small angles of the wave mixing geometry, which minimizes beam walk-off effects and enables better depth resolution and wider field of views, (ii) they have shown strong phase conjugate reflectivities (which gives good imaging sensitivities) for low intensities of cw laser radiation; and (iii) they are inexpensive; and (iv) their optical nonlinearities can be significantly improved by the application of an external electric field.

## D. Plans For Involvement And Interaction With Federal Laboratories

Non-destructive, non-invasive characterization of the porous structure of transparent, but highly diffusive, epoxy composites is an important Air Force problem. Characterization of polymers and thin film coatings used for Air Force components and systems is also a critical need. We have received strong interest in this research from a number of DoD groups with whom we plan to collaborate.

Mary J. Miller, Team Chief, Nonlinear Optical Processes Team, ARL, has expressed very strong interest in collaborating with us in this research program in numerous E-Mail messages that we have exchanged (she has also written a *letter of support*, enclosed at the end of the proposal package). Our collaboration plans involve combining the ARL research expertise in imaging based on 2-wave mixing in photorefractives with our expertise in imaging based on DFWM in thin liquid crystal films. We have had a number of successful collaborations with this ARL group in the past in nonlinear optics and power limiting, and we look forward to some very successful interactions with them in the proposed imaging program.

We have also communicated some of our ideas with Bob Crane at Wright Labs (who is familiar with the Air-Force non-destructive evaluation issues), and he said "I must confess that I have had very similar ideas, but have not pursued these to any reasonable degree. For this reason I would be interested in what you have learned and what you propose." His research involves the many problems associated with the problems of assessing damage in very old aircraft. These structures will be extensively repaired and reworked in the future; methods to assess damage in terms of cracking or corrosion are high on his list of things that must be done soon. In addition, the development of techniques to interrogate repaired structures to assess their structural reliability, and he

said that "any ideas that you have in these areas would be of interest". He suggested that "I would ask you to stick to the generic problem because I have not listed the multitude of engineering constraints that any solution will face. We in the AF Labs will address this aspect of the problem and lease the science to you". In the proposed program, we plan to maintain an active collaboration with Dr. Crane and to periodically send him write-ups of our technical results and publications. We plan to also visit him to obtain input from him on specific Air Force non-destructive evaluation issues, so that our research effort (which will concentrate on basic optical science) will be relevant to Air Force non-destructive evaluation problems.

## E. Education of Graduate Students and Post-Doctoral Research Associates

Dr. Lawson has an active research group that involves postdoctoral, graduate, and undergraduate students from Physics, and the joint University of Alabama Materials Science Graduate Programs. The proposed research effort will provide a cross-disciplinary educational experience in optical imaging, nonlinear optics, optical wave mixing, and materials characterization. One post-doctoral research associate and two graduate students will be directly supported by the proposed grant, and a third and fourth graduate student will also be directly involved in the research, supported by other fellowships (one by a Graduate Assistant Fellowship and the other by a Comprehensive Minority Faculty Development Fellowship).

The number of graduate students working with NLO materials may enable the investigators to offer a special topics course in this area. In addition, a number of undergraduate students will also participate in the project through the NSF-funded Alabama Alliance for Minority Participation (AAMP) Program, the NSF-REU Summer Research Program in Experimental and Computational Materials Research, and Physics Department senior research projects.

## F. Experimental Facilities

The proposed imaging research will be performed in the NLO/Laser Research Laboratory and the UAB Optical Imaging / Fiber Optics Laboratory. These laboratories have equipment detailed below.

UAB's Optical Imaging / Fiber Optics Laboratory has been outfitted in the last three years with approximately 100K of start-up funds for Dr. Lawson and approximately 240K of grant funds from NSF and NASA for projects that Dr. Lawson is leading. This laboratory contains an argon laser-pumped Ti:sapphire laser which provides ultra-short laser pulses for coherence-based imaging applications. The laboratory also is well stocked with numerous high coherence and wide spectrum diode light sources, numerous detector systems, a wide variety of fiber optics components and support equipment, and computer-controlled scanning systems.

UAB's Nonlinear Optics and Laser Laboratory has been outfitted in the last two years with approximately 230K of internal UAB funds and 780K of funds from a current ARO grant (Dr. Lawson is PI) and two recently concluded NSF grants of which Dr.

Lawson was PI. This laboratory houses state-of-the art spectroscopic analysis systems including an Acton Research Corporation spectrometer system, a Shimadzu spectrophotometer system, and an imaging polychromator - lens-coupled intensified CCD camera combination. Auxiliary equipment includes a 80 ps resolution Tektronics digitizer, a Stanford Research boxcar integrator, a Janis crystrostat, a Spiricon beam profiling system, and a wide variety of optical components and mounts. In addition to a low power Nd:YAG laser, the laboratory also houses a Spectra-Physics GCR-250 high power Nd:YAG laser with injection seeder for single longitudinal mode. This GCR-250 laser is also used as a pump for a "MALSAN" wavelength tunable LiF color center laser (820-1270, 420-625 and 270-312 nm), which is used for the wavelength tunable DFWM and Z-scan experiments described in this proposal. A Light Age alexandrite laser can be tuned in wavelength from 720 nm to 800 nm, extended to 300-1500 nm with our Raman cell module and frequency doublers and triplers. The temporal pulse width of the laser can be varied from 100 ps to 200 ms. More recently, a picosecond mode-locked Nd:YAG laser has been ordered to provide an additional ps pulsed laser source for optical measurements.

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**CHRISTOPHER M. LAWSON**, Associate Professor, Physics, University of Alabama at Birmingham (UAB), 1300 University Blvd., Birmingham, AL 35294-1170, (205)975-5059; INTERNET: lawson@phy.uab.edu

**EDUCATION:** Ph.D. Physics, 1981, Oklahoma State University (B.S. Physics, 1976), M.S. Physics, University of Colorado at Boulder, 1979

**PH.D. THESIS ADVISOR:** Richard Powell, Director, Univ. Arizona Opt. Sci. Cntr.

**PROFESSIONAL EXPERIENCE:**

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| 1993-Present | Associate Professor, Physics, University of Alabama at Birmingham |
| 1991-1993 | Principle Scientist, Optical Sciences, BDM International |
| 1989-1991 | Director, Optical Sciences, BDM International |
| 1985-1989 | Manager, Optical Sciences, BDM International |
| 1984-1985 | Scientist, Optics and Information Processing, BDM International |
| 1981-1984 | Staff Scientist, Fiber Optics, Gould Research Labs, Gould, Inc. |

**RESEARCH ACTIVITIES**

Dr. Lawson's research interests, with short term support by NSF and NASA, involve the development of optical sensing techniques for medical imaging and materials processing characterization. Recent research has involved the use of nonlinear wave mixing techniques for single pulse coherence-based depth-resolved 2-D imaging. Also has used optical low coherence interferometry for non-invasive characterization of semiconductor (solar silicon) growth processes and has recently used similar techniques to image protein crystals in solution. Before coming to UAB he studied the use of optical time domain reflectometry for materials defect characterization. Earlier still, Dr. Lawson developed fiber optical sensors for such diverse applications as measurement of material structural properties, stress/strain, electric fields, temperature, pressure, and sound.

Dr. Lawson's other research interests, with long term support by Army Research Office, are directed toward the development and characterization of nonlinear optical (NLO) materials for optical switching and power limiting applications. He currently studies NLO processes in metal-organic complexes using Z-scan and degenerate four wave mixing (DFWM) spectroscopy. Before coming to UAB, he performed NLO research as the PI of a long-term DARPA program involving theoretical and experimental investigations of NLO absorption, NLO plasma scattering, and cavitation/total internal reflection (TIR) mechanisms in carbon microparticle suspensions for power limiting applications. Previously, he studied the NLO materials properties of photorefractive materials.

**TEACHING ACTIVITIES**

Has taught numerous Introductory College Physics classes, and has developed and taught a 3 part Optics curricula, covering paraxial optics, matrix theory, computer optical design (using CODE-V optical design software), interferometry, coherence, diffraction, lasers, polarization, electro-optics, anisotropic optics, and nonlinear optics. Currently directs research of four graduate students and one post-doctoral research associate.

## FIVE RECENT OPTICAL IMAGING PUBLICATIONS

1. V. Fleurov, D. Brown, and C. Lawson, "Low-Coherence Reflectometry Through Scattering Media With Degenerate Four-Wave Mixing In A Thin Nonlinear Optical Layer", accepted by the Journal of Nonlinear Optics.
2. C.M. Lawson, D.A. Forrestall, T. Zhai, M. Shen, and L.J. DeLucas, "Three-Dimensional Imaging Of Protein Crystals Using Optical Low Coherence Reflectometry", Microwave & Optical Technology Letters Vol. **13**, pp. 63-66, (1996).
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## FIVE RECENT NONLINEAR OPTICS PUBLICATIONS

6. G. Gray & C. Lawson, "Structure-Property Relationships In Transition Metal-Organic 3rd-Order NLO Materials", Chapter 10 in ***Opto-Electronic Properties of Inorganic Compounds***, M. Roundhill & J. Fackler, ed., (Plenum Publ., N.Y., N.Y.). (In Press)
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1. PI, "New Metal-Organic Nonlinear Optical Complexes", 9/15/96-9/14/99, ARO Grant DAAH04-96-1-0400 for **$329,652**.
2. Task Leader, "Fiber Optic Low Coherence Interferometry for Protein Crystal Growth Monitoring" task for **$113,273**, 10/1/96-9/30/97, part of NASA Space Station Grant.
3. Co-PI (John Dimmock at UAH is PI), "Alabama Consortium for Optical Technology", 10/1/95-9/30/97, **$80,802,** NSF Grant OSR-9553348.
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5. PI, "AL Laser Research Initiative", 7/94-7/96, NSF Grant OSR-9450570 **$199,991**.
6. PI / Consultant, "Fiber Optic Sensor Development For Silicon Growth Monitoring" 10/92-10/93; Mobil Solar Energy Corporation, Contract ZM-2-11040-3 for **$159,327**.
7. PI, "Nonlinear Optics For Hybrid Optical Power Limiter Development", 2/89-12/92, DARPA Contract DAAB07-89-C-F401 for **$1,182,000.**